

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
27 December 2001 (27.12.2001)

PCT

(10) International Publication Number
WO 01/97957 A1

(51) International Patent Classification⁷: **B01D 71/06**

(21) International Application Number: PCT/KR01/01076

(22) International Filing Date: 22 June 2001 (22.06.2001)

(25) Filing Language: Korean

(26) Publication Language: English

(30) Priority Data:
2000/34948 23 June 2000 (23.06.2000) KR

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(81) Designated States (*national*): CN, JP, US.

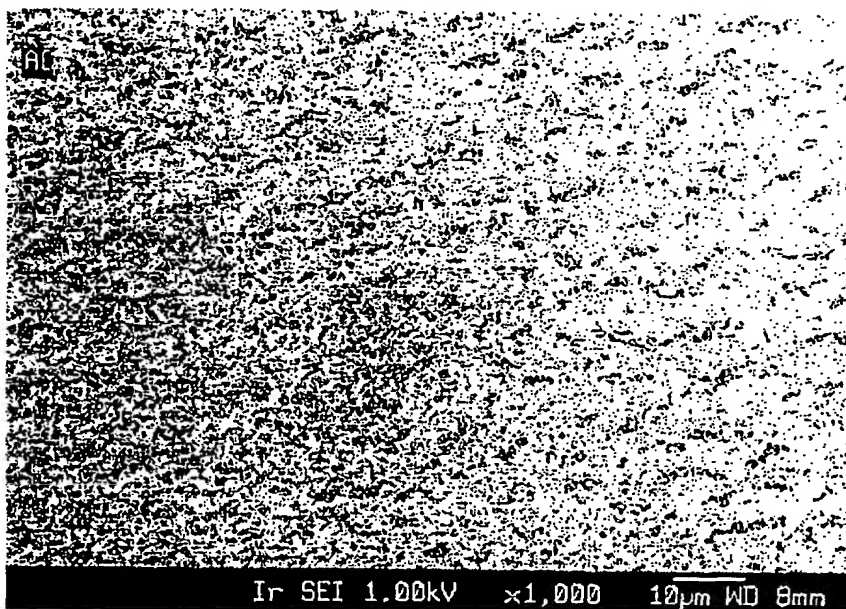
(84) Designated States (*regional*): European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE, TR).

Published:

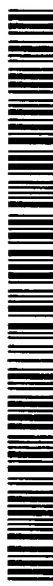
- with international search report
- before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: MULTI-COMPONENT COMPOSITE MEMBRANE AND METHOD FOR PREPARING THE SAME



(57) Abstract: The present invention relates to a multi-component composite separate membrane and method for preparing the same, and it provides a composite membrane comprising active layers and support layers, wherein the support layers are located on the interior side of the membrane and the active layers are located on the exterior side of the membrane.



WO 01/97957 A1

THE USE BLANK (USPTO)

MULTI-COMPONENT COMPOSITE MEMBRANE AND METHOD FOR PREPARING THE SAME

CROSS REFERENCE TO RELATED APPLICATION

5 This application is based on application No. 2000-34948 filed in the Korean Industrial Property Office on June 23, 2000, the content of which is incorporated hereinto by reference.

BACKGROUND OF THE INVENTION

(a) Field of the Invention

10 The present invention relates to a multi-component composite membrane and a method for preparing the same, and more particularly, to a multi-component composite membrane comprising a support layer and an active layer having a structure as dense as the conventional active layers, which is capable of having pores formed thereon according to process
15 conditions and with improved permeability due to the pores of a controllable size, the composite membrane having characteristics of an active layer, and with an interfacial adhesion strength between the support layer and the active layer strengthened by ion beams irradiation, and a method of preparing the same.

20 (b) Description of the Related Art

Many types of membranes are currently in use, such as microfiltration membranes, ultrafiltration membranes, gas separation membranes, pervaporation membranes, and reverse osmosis membranes.

The present invention relates to a microfiltration membrane, and in particular to a separator comprising polyolefins such as polyethylene and
25 polypropylene, for a rechargeable lithium ion battery.

As one of the polyolefins, when high crystalline polypropylene (HCPP) is used for a separator of the present invention, it is expected that the permeability of the separator will increase. The crystallinity of common

polypropylenes is less than 50%, but the crystallinity of HCPP is greater than 50% and it is highly isotactic, so that density, melting point, heat of fusion, and crystallization temperature are high, and characteristics such as rigidity, heat-resistance, impact strength, scratch-resistance, and dimensional stability are excellent.

A composite membrane is generally prepared by interfacial polymerization, modification of membranes, and dip coating. Dip coating is widely used in order to prepare the composite membrane, by using a microporous membrane such as a microfiltration membrane or an ultrafiltration membrane as a support layer, coating the microporous membrane with a solution of a material used as an active layer, and drying the coated membrane. The composite membrane prepared by dip coating has a support layer comprising regularly-sized pores, and an active layer having a dense structure with few pores. The composite membrane is limited in application, since the active layer has few pores of a size similar to those of the microfiltration or ultrafiltration membranes, and it is easily delaminated due to a weak interfacial adhesion strength between the support layer and the active layer.

The composite membrane may be prepared by coating a polymer on the microporous membrane as disclosed in U.S. Patent Nos. 3,249,109, 4,388,189, and 5,102,552. In addition, a hydrophilic monomer, such as an acrylic acid, and polymers such as polyethylene oxide are grafted with corona treatment so that the membrane has a modified surface, and in particular so that it has hydrophilicity as disclosed in U.S. Patent Nos. 4,346,142, 5,085,775, and 5,294,346. However, though the membrane has a modified surface and hydrophilicity, the method of graft polymerization is not applied, since the process is complicated and permeability of the membrane is not satisfactory.

A separator having regularly-sized pores for a common battery is coated with a polymer electrolyte solution, and it is used as a separator for a rechargeable lithium ion battery as disclosed in U.S. Patent No. 5,716,421 and European Patent No. 0933824A2. However, when the separator is prepared by the aforementioned method, the membrane has a dense structure, that is,

no pores are formed on the surface of the membrane, and permeability (e.g. air permeability) deteriorates, and the interfacial adhesion strength between the support layer and the active layer is inadequate.

SUMMARY OF THE INVENTION

5 It is an object of the present invention to provide a multi-component composite membrane comprising a support layer and an active layer having a structure as dense as the conventional active layers, which is capable of having pores formed thereon according to process conditions and with improved permeability due to the pores of a controllable size, the composite membrane
10 having characteristics of an active layer, and with an interfacial adhesion strength between the support layer and the active layer strengthened by ion beams irradiation, and a method of preparing the same.

In order to accomplish the object, the present invention provides a multi-component composite membrane comprising a support layer and two
15 active layers.

Furthermore, the present invention provides a preparation method of a multi-component composite membrane comprising the steps of:

- a) preparing a precursor film by injection of a polymer, which is used for a support layer, into an extruder;
- 20 b) annealing the precursor film at a temperature less than a melting point of the polymer;
- c) irradiating ion beams on either or both surfaces of the annealed precursor film with the help of an reactive gas;
- d) coating both surfaces of the irradiated precursor film with a polymer
25 solution, which is used for an active layer;
- e) drying the coated precursor film;
- f) low temperature-stretching the dried precursor film at a temperature less than a room temperature;
- g) high temperature-stretching the low temperature-stretched
30 precursor film at a temperature less than a melting point of the polymer; and

- h) heat setting the high temperature-stretched precursor film with a tension at a temperature less than the melting point of the polymer.

BRIEF DESCRIPTION OF THE DRAWINGS

A more complete appreciation of the invention, and many of the attendant advantages thereof, will be readily apparent as the same becomes better understood by reference to the following detailed description when considered in conjunction with the accompanying drawings, wherein:

FIG. 1 is a scanning electron microscope (SEM) photograph showing a surface of a composite membrane of Example 1 according to the present invention; and

FIG. 2 is a SEM photograph showing a surface of a conventional composite membrane according to Comparative Example 1.

DETAILED DESCRIPTION OF THE INVENTION

In the following detailed description, only the preferred embodiment of the invention has been shown and described, simply by way of illustration of the best mode contemplated by the inventors of carrying out the invention. As will be realized, the invention is capable of modification in various obvious respects, all without departing from the invention. Accordingly, the drawings and description are to be regarded as illustrative in nature, and not restrictive.

The present invention is described in detail as follows.

The present invention provides a composite membrane and a preparation method for the same that involves coating a common film having no pores with an active material, instead of as in the conventional method of coating a microporous film with an active material.

The composite membrane of the present invention is prepared by joint-use of a conventional dry process in which the pores are formed by stretching, and a phase inversion that is used with a solution. In addition, when an ion beam irradiation step is added in the preparation steps to prepare the membrane of the present invention, an interface bond between a support layer and an active layer is improved.

The preparation method using a conventional dry process is a method in which pores are formed by rupturing a relatively weak amorphous region through cold stretching after orientating a polymer crystalline region in a certain direction, and the orientation uniformity of the crystalline region is critical for characteristics of the membrane.

The method using phase inversion is a method in which pores are formed by phase-separation of a polymer and a solvent from the solution under a controlled temperature, or the use of a non-solvent after preparing a polymer solution.

In order to modify the surface, an ion beam irradiation process is used in which gases such as gaseous argon, hydrogen, oxygen, nitrogen, and carbon dioxide are ionized and irradiated to the surface under an atmosphere of reactive gases to be reacted with the ions and the surface when the ionized gases collide with the surface of the membrane.

In the present invention, in order to prepare a material used as a support layer, a precursor film is prepared in one step of the drying process, it is coated with a polymer solution used for an active layer, it is phase-separated from the polymer solution under suitable conditions, and it is stretched, and thereby the membrane is prepared and pores are formed on the membrane.

During the membrane preparation, in order to increase an interfacial adhesion strength between the support layer and the active layer, the ion beam irradiation process is performed before the coating process, so that the membrane surface is modified. The composite membrane of the present invention comprises materials having pores, which are used for the support layer and the active layer, respectively. The pore size and distribution of the support layer and active layer are different from each other, with the pores of the support layer being formed by a stretching process after orientating a polymer crystalline region in a certain direction during the precursor film preparation. The pores of the active layer, on the other hand, are formed by a stretching process after forming a densely structured polymer film through phase-inversion. Micro-cracks and micro-pores of the polymer film can be

formed according to the phase-inversion conditions before the film is stretched, so the degree of pore formation is controllable according to said phase-inversion conditions.

The support layer of the present invention has the same characteristics as a membrane prepared from the conventional dry process, and the active layer has pores with various sizes according to the process conditions. In addition, inter-diffusion among the polymer chains of the support layer and active layer improves through high temperature-stretching and heat-setting, and the surface bond between the support layer and the active layer strengthens. When ion beams are irradiated to the layers, the surface bond may further strengthen.

The material used for the support layer of the present invention is not limited to a certain material, and it generally includes one or more materials selected from the group consisting of high density polyethylene, low density polyethylene, linear low density polyethylene, polypropylene, high crystalline polypropylene, polyethylene-propylene copolymer, polyethylene-butylene copolymer, polyethylene-hexene copolymer, polyethylene-octene copolymer, polystyrene-ethylene-butylene-styrene copolymer, polystyrene, polyphenylene oxide, polysulfone, polycarbonate, polyester, polyamide, polyurethane, polyacrylate, polyvinylidene chloride, polyvinylidene fluoride, polysiloxane, polyolefin, ionomer, polymethylpentene, and hydrogenated oligocyclopentadiene (HOCPP), and a mixture thereof, and preferably only material, blended material, or laminated material selected from the aforementioned group is used.

The polymer of the polymer solution used for the active layer is selected according to the eventual use of the composite membrane, and it preferably includes at least one material selected from the group consisting of polyethylene, polypropylene, polyvinylidene fluoride, polyvinylidene fluoride-hexafluoropropylene copolymer, polyethylene oxide, polypropylene oxide, polybutylene oxide, polyurethane, polyacrylonitrile, polyacrylate, polyacrylic acid, polyamide, polyacrylamide, polyvinylacetate, polyvinylpyrrolidone,

polytetraethylene glycol diacrylate, polysulfone, polyphenylene, oxide, polycarbonate, polyester, polyvinylidene chloride, polysiloxane, and a polyolefin inomer, and a derivative thereof.

The solvent of the polymer solution is selected according to the polymer used, and it preferably includes at least one solvent selected from 1-methyl-2-pyrrolidone (NMP), acetone, ethanol, n-propanol, n-butanol, n-hexane, cyclohexanol, acetic acid, ethyl acetate, diethyl ether, dimethyl formamide (DMF), dimethylacetamide (DMAc), dioxane, tetrahydrofuran (THF), dimethyl sulfoxide (DMSO), cyclohexane, benzene, toluene, xylene, and water, and a mixture thereof.

The polymer solution is preferably used under the following conditions. During the coating process of the polymer solution, a common film having no pores is dip-coated in the polymer solution, with the concentration of the polymer solution preferably being 0.01 wt% or greater. In addition, it is preferable that the drying of the coated polymer is performed at a relative humidity ranging from 1 to 100% under an atmosphere of a gas selected from gases comprising nitrogen, oxygen, carbon dioxide, and air at a saturated vapor pressure of less than a saturated vapor pressure of the solvent. The thickness of the active layer after coating and drying preferably ranges from 0.1 to 20 μm .

The ion beam irradiation is performed under a vacuum ranging from 10^{-1} to 10^{-6} torr, with activated electrons, hydrogen, helium, oxygen, nitrogen, carbon dioxide, air, fluorine, neon, argon, krypton, and N_2O , and a mixture thereof, the aforementioned ion particles having an energy ranging from 0.01 to 10^6 keV. Preferably, the amount of the ion particle ranges from 10^5 to 10^{20} ions/ cm^2 . The reactive gases preferably include helium, hydrogen, oxygen, nitrogen, ammonia, carbon monoxide, carbon dioxide, chlorofluorocarbon, methane, and N_2O , and a mixture thereof, and the flow rate of the reactive gases preferably ranges from 0.5 to 20 ml/minute.

The preparation method of the composite membrane according to the present invention comprises the following steps.

a) Precursor film preparation of the support layer: A precursor film is

prepared by extrusion of a polymer used for a support layer with an extruder equipped with a T-die or tubular die.

- 5 b) Annealing: The precursor film is annealed in a dry oven at a temperature lower than a melting point of the polymer so that the precursor film has an increased crystallinity and ratio of elastic recovery.
- 10 c) Irradiation of ion beams with the help of an reactive gas: After the precursor film is placed in a vacuum chamber and ionized gas is injected into an ion gun so that the gas has an energy, the ion particles having an energy are irradiated on either or both sides of the precursor film, depending on various currents. A power source is controlled in order that the energy of the ions ranges from 0.01 to 10^6 keV. While irradiating the ion beams, an reactive gas of which flow rate is varied from 0.5 to 20 ml/minute is injected into the vacuum chamber in order to modify a surface of the precursor film. The modification of the surface of the precursor film may be performed before or after annealing, depending on desired physical properties of the multi-component composite membrane.
- 15 d) Coating the precursor film with a polymer solution used for an active layer: After a polymer solution is prepared by dissolving a polymer used for an active layer in a desired solvent, the precursor film is coated with the polymer solution. The precursor film may be used before or after annealing. In addition, before coating, the irradiation of ion beams with the help of the reactive gas may be performed, depending on the physical properties of the multi-component composite membrane. Concentration and coating conditions may be varied according to materials used and the eventual use of the composite membrane.
- 20 e) Formation of a polymer film by phase-inversion: After coating, the solvent is vaporized under suitable conditions. The structure of the polymer film of the active layer depends on the drying conditions.
- 25
- 30

- f) Low-temperature stretching: Microcracks are formed by mono-axially stretching the annealed film with use of a roll or other different stretching machines at a temperature lower than room temperature.
- 5 g) High-temperature stretching: Micropores having desirable sizes are formed and mechanical properties are provided to a membrane by the ion-beam-irradiation and mono-axially or bi-axially stretching the low temperature stretched film with use of a roll or other machine at a temperature lower than a melting point of the polymer of the support and active layers.
- 10 h) Heat-setting: After the high-temperature stretching, the film is heat-set at a temperature lower than a melting point of the polymer of the support and active layers under tension, for a certain time.

The preparation steps of the multi-component composite membrane describe the overall processes for preparing a membrane having optimum physical properties, wherein the membrane can be prepared not only by skipping some of the steps or adding processes depending on physical properties, but also by changing the sequence of each of the steps.

The following Examples and Comparative Examples illustrate the present invention in further detail, but the present invention is not limited by these examples.

The microporous membranes prepared by the following Examples and Comparative Examples were evaluated according to the following characteristics:

- 25 a) thickness;
- b) air permeability: JIS P8117;
- c) pore size: scanning electron microscope (SEM), transmission electron microscope (TEM);
- d) interfacial adhesion strength: JIS Z0237; and
- 30 e) a wet-out rate of electrolyte (electrolyte used: ethylene carbonate (EC):dimethyl carbonate (DC) = 4:6)

Example 1: Composite membrane prepared from high crystalline polypropylene and Kynar461

A high crystalline polypropylene was used for a support layer and a polyvinylidene fluoride (PVDF) was used for an active layer in order to prepare a precursor film, and the precursor film was stretched through a dry process in order to prepare a composite membrane.

(Preparation of a precursor film)

High crystalline polypropylene was used for a component of a support layer. It has a melting index of 0.50 g/min, a density of 0.91 g/cc, a melting point of 166.5°C measured with a dynamic scanning calorimeter (DSC), a crystallization temperature of 134.5°C, a crystallinity of 57%, isotacticity of 98% measured by C¹³ nuclear magnetic resonance (NMR), and an atactic fraction of about 2% measured after dissolution in xylene, and a precursor film was prepared from the high crystalline polypropylene with use of a single screw extruder equipped with T-die and a take-up device. Extrusion temperature and cooling-roll temperature was 220°C and 80°C respectively, take-up speed was 20 m/min, and a draw down rate (DDR) was 60.

(Annealing)

The prepared precursor film was annealed in a dry oven at 150°C for 1 hour.

(Coating)

After annealing, a solution prepared by dissolving Kynar461 (a product by Elf Atochem North America Co.) having low crystallinity in acetone was dip-coated on both sides of the prepared precursor film. The coating was performed under air while maintaining 60% relative humidity, and the acetone was vaporized at the same condition of 60% relative humidity. Thickness of the coated Kynar461 was about 3 μm .

(Low-temperature stretching)

After coating, the coated film was mono-axially low-temperature stretched to 50% of the stretching ratio based on the initial length of the film at room temperature.

(High-temperature stretching)

After low-temperature stretching, the low-temperature-stretched film was mono-axially high-temperature stretched to 100% of the stretching ratio based on the initial length of the film, at 140°C.

5 (Heat-setting)

After the high-temperature stretched film was heat-set at 140°C. under tension for 10 minutes, a composite membrane was prepared by cooling the heat-set film.

Properties of the composite membrane are represented in Table 1.

10 It is shown in Table 1 that micropores form on an active layer as well as a support layer according to Example 1, and air permeability of the membrane according to Example 1 improves, compared to no micropores forming on the Kynar461 used for an active layer (See Fig1. and Fig. 2) when the Kynar461 was coated on a separator as in the conventional preparation method. In
15 addition, the interfacial adhesion strength between the support layer and the active layer increased. It is supposed that the wet-out rate of electrolyte increased due to the change of morphology and the increase of adhesion strength.

20 Example 2: A composite membrane prepared from high crystallinity polypropylene and Kynar461 with irradiation of ion beams

A composite membrane was prepared by same method of Example 1, except that ion beams were irradiated on a precursor film before coating with the Kynar461 solution. After the precursor film prepared in the same manner as in Example 1 was placed in a vacuum chamber while keeping the pressure
25 ranging from 10^{-5} to 10^{-6} torr, argon cations were irradiated to both sides of the precursor film with an ion gun, and simultaneously oxygen used as an reactive gas was injected into the chamber in an amount of 4 ml/min in order to treat the precursor film surface. Energy of the ion beams was 0.5 keV, and the irradiation amount of ions was 10^{16} ions/cm². After the ion-beam irradiation, a
30 composite membrane was prepared in the same manner as in Example 1.

In Table 1, it is shown that pores were formed on both the support layer

and the active layer as in Example 1, and in particular, the interfacial adhesion strength between the support layer and the active layer, and the wet-out rate of the electrolyte were appreciably improved.

5 Example 3: A membrane prepared from high density polyethylene/
Kynar461.

A composite membrane was prepared in the same manner as in Example 1, except that high density polyethylene was used for a support layer instead of high crystalline polypropylene. The high density polyethylene had a melt index of 0.3 g/10 min and a density of 0.964 g/cc. A precursor film was
10 prepared in the same manner as in Example 1. The extrusion temperature and cooling-roll temperature of the take-up device were respectively 200°C and 80°C, the take-up speed of the film was 30 m/min, and the draw-down ratio of the prepared precursor film was 80. The prepared precursor film was annealed in a dry oven at 125°C for 1 hour. Both sides of the annealed
15 precursor film were coated with Kynar461 in the same manner as in Example 1. The coated precursor film was mono-axially stretched at room temperature to 50% of the stretching ratio based on the initial length of the film, and then it was immediately mono-axially high-temperature stretched to 50% of the stretching ratio based on the initial length of the film, at 120°C. The high-temperature
20 stretched film was heat-set at 120°C under tension for 10 minutes, and then a composite membrane was prepared by cooling the heat-set film. Table 1 shows properties of the prepared composite membrane.

In Table 1, it is observed that pores formed on both the support layer and the active layer as in Example 1, and the interfacial adhesion strength and
25 wet-out rate of the electrolyte improved.

Comparative Example 1: A composite membrane prepared from
Celgard2400 and Kynar461

An active layer was coated on a microporous membrane by the conventional method.

30 Celgard2400 (a product by Celanese Co.) prepared from only polypropylene was used for the porous membrane as a support layer, Kynar461

was used for an active layer as in Examples 1, 2 and 3, and the Kynar461 solution was coated on the Celgard2400 having pores, and thereby a composite membrane was prepared.

Figs. 1 and 2 show that the composite membrane of Comparative Example 1 has no pores, unlike the composite membrane of the examples according to the present invention that have pores formed on the active layer.

Table 1 shows properties of the prepared composite membranes. It is shown that the composite membrane prepared from Celgard2400 and Kynar461 had an air permeability too inferior to measure, and the interfacial adhesion strength and wet-out rate of electrolyte were inferior.

Table 1

		Example 1	Example 2	Example 3	Comparative Example 1
Thickness (μm)		20	20	20	20
Pore size (μm)	Support layer	0.3 x 0.1	0.3 x 0.1	0.4 x 0.1	0.3 x 0.1
	Active layer	0.8 x 0.3	0.8 x 0.3	0.6 x 0.3	Unable to be measured
Air permeability (sec/ 100 cc)		560	565	620	Unable to be measured
Interfacial adhesion strength (gf)		180	250	240	85
Wet-out rate of an electrolyte (sec)		10	8	9	45

The composite membrane prepared by the conventional method has a permeability that is too inferior to be measured, but the composite membranes of the present invention have an improved air permeability ranging from 560 to 620 sec/ 100 cc, because both the active layer and the support layer have a dense structure with pores of a controllable size prepared under suitable preparation conditions. In addition, the active layer located on the exterior side of the composite membrane has good properties. That is, the support layer of

the present invention has the same properties as the membrane prepared by the conventional dry process, and the active layer has pores with various sizes according to the process condition.

In addition, while the composite membrane prepared by the conventional method has an interfacial adhesion strength of 85 gf, the composite membrane of the present invention has an improved interfacial adhesion strength ranging from 180 to 250 gf. The improved interfacial adhesion strength results from high-temperature stretching and heat-setting, that is, the interfacial adhesion strength increases because the mutual bond between polymer chains of the support and active layers strengthens. The interfacial adhesion strength improves further by irradiation of ion beams.

Furthermore, the wet-out rate improves appreciably, and it is supposed that the improvement of the wet-out rate is due to changes of morphology and an increase of the interfacial adhesion strength.

While the present invention has been described in detail with reference to the preferred embodiments, those skilled in the art will appreciate that various modifications and substitutions can be made thereto without departing from the spirit and scope of the present invention as set forth in the appended claims.

WHAT IS CLAIMED IS:

1. A multi-component composite membrane comprising active layers and support layers, wherein a support layer is located between active layers.
- 5 2. The multi-component composite membrane according to claim 1, wherein a component of a support layer is at least one polymer selected from the group consisting of high density polyethylene, low density polyethylene, linear low density polyethylene, polypropylene, high crystalline polypropylene, polyethylene-propylene copolymer, polyethylene-butylene copolymer,
10 polyethylene-hexene copolymer, polyethylene-octene copolymer, polystyrene-butylene-styrene copolymer, polystyrene-ethylene-butylene-styrene copolymer, polystyrene, polyphenylene oxide, polysulfone, polycarbonate, polyester, polyamide, polyurethane, polyacrylate, polyvinylidene chloride, polyvinylidene fluoride, polysiloxane, polyolefin, ionomer, polymethylpentene, and
15 hydrogenated oligocyclopentadiene (HOCP), and a derivative thereof.
3. The multi-component composite membrane according to claim 2, wherein the high crystalline polypropylene has one or more physical properties selected from the group consisting of a density of 0.905 g/cc or more, a melting point of 164°C or higher, a crystallization temperature of 125°C or
20 higher, a crystallinity of 50% or greater, an isotacticity of 96% or greater, and an atactic fraction of 5% or less.
4. The multi-component composite membrane according to claim 1, wherein a pore size of a support layer ranges from 0.001 to 10 μm .
5. The multi-component composite membrane according to claim
25 1, wherein a thickness of a support layer ranges from 1 to 50 μm .
6. The multi-component composite membrane according to claim 1, wherein a component of the active layers is one or more polymers selected from the group consisting of polyethylene, polypropylene, polyvinylidene fluoride, polyvinylidene fluoride-hexafluoropropylene copolymer, polyethylene oxide, polypropylene oxide, polybutylene oxide, polyurethane, polyacrylonitrile,
30 polyacrylate, polyacrylic acid, polyamide, polyacrylamide, polyvinylacetate,

polyvinylpyrrolidone, polytetraethylene glycol diacrylate, polysulfone, polyphenylene oxide, polycarbonate, polyester, polyvinylidene chloride, polysiloxane, and polyolefin ionomer, and a derivative thereof.

7. The multi-component composite membrane according to claim 5 6, wherein a solvent for the component of the active layers is one or more solvents selected from the group consisting of 1-methyl-2-pyrrolidone (NMP), acetone, ethanol, n-propanol, n-butanol, n-hexane, cyclohexanol, acetic acid, ethyl acetate, diethyl ether, dimethyl formamide (DMF), dimethylacetamide (DMAc), dioxane, tetrahydrofuran (THF), dimethyl sulfoxide (DMSO), 10 cyclohexane, benzene, toluene, xylene, and water, and a mixture thereof.

8. The multi-component composite membrane according to claim 1, wherein a pore size of an active layer is equal to or less than 10 μm .

9. The multi-component composite membrane according to claim 1, wherein a thickness of an active layer ranges from 0.01 to 20 μm .

10. The multi-component composite membrane according to claim 15 1, wherein air permeability of the composite membrane is equal to or less than 7,000 sec/100 cc.

11. The multi-component composite membrane according to claim 1, wherein a wet-out rate of the composite membrane is equal to or less than 20 30 seconds.

12. The multi-component composite membrane according to claim 1, wherein the composite membrane comprises two or more active layers and one or more support layers, and the composite membrane has a structure of triple or multiple layers.

13. The multi-component composite membrane according to claim 25 1, wherein a support layer is blended or laminated with the polymer component of claim 2.

14. The multi-component composite membrane according to claim 1, wherein the composite membrane is used for water treatment, hemodialysis, 30 enzyme purification, patches for drug delivery, gas separation, pervaporation, reverse osmosis, or electrolysis separation.

15. A separator for batteries comprising the multi-component composite membrane of claim 1.

16. A rechargeable lithium ion battery or a rechargeable lithium ion polymer battery comprising the multi-component composite membrane of claim 1 as a separator.

17. A preparation method of a multi-component composite membrane comprising steps of:

- a) preparing a precursor film by injection of a polymer, which is used, for a support layer into an extruder;
- b) annealing the precursor film at a temperature less than a melting point of the polymer;
- c) irradiating ion beams on either or both surfaces of the annealed precursor film;
- d) coating both surfaces of the irradiated precursor film with a polymer solution, which is used for an active layer, with the help of an reactive gas;
- e) drying the coated precursor film;
- f) low temperature-stretching the dried precursor film at a temperature less than room temperature;
- g) high temperature-stretching the low temperature-stretched precursor film at a temperature less than a melting point of the polymer; and
- h) heat-setting the high temperature-stretched precursor film under tension at a temperature less than the melting point of the polymer.

18. The preparation method according to claim 17, wherein the polymer solution of step d) is coated on both sides of the precursor film by dip-coating.

19. The preparation method according to claim 17, wherein a concentration of the polymer solution of step d) is equal to or greater than 0.01 wt%.

20. The preparation method according to claim 17, wherein the drying of step e) is performed at a relative humidity ranging from 1 to 100%.

21. The preparation method according to claim 17, wherein the drying of step e) is performed under saturated vapor pressure.

5 22. The preparation method according to claim 17, wherein the drying of step e) is performed under a gas atmosphere selected from the group consisting of nitrogen, oxygen, carbon dioxide, and air atmosphere.

23. The preparation method according to claim 17, wherein an active layer having a thickness in the range of 0.1 to 20 μm is formed through
10 the coating and drying of steps d) and e).

24. The preparation method according to claim 17, wherein the irradiating of ion beams of step c) is performed under a vacuum ranging from 10^{-1} to 10^{-6} torr.

25. The preparation method according to claim 17, wherein the
15 irradiating of ion beams of step c) is performed by activation of electrons and a gas selected from the group consisting of hydrogen, helium, oxygen, nitrogen, carbon dioxide, air, fluorine, neon, argon, krypton, N_2O , and a mixture thereof such that the gas has an energy ranging from 0.01 to 10^6 keV; and irradiation of the ion beams on the surface of the precursor film.

20 26. The preparation method according to claim 17, wherein an irradiation amount of the ion beams ranges from 10^5 to 10^{20} ions/ cm^2 .

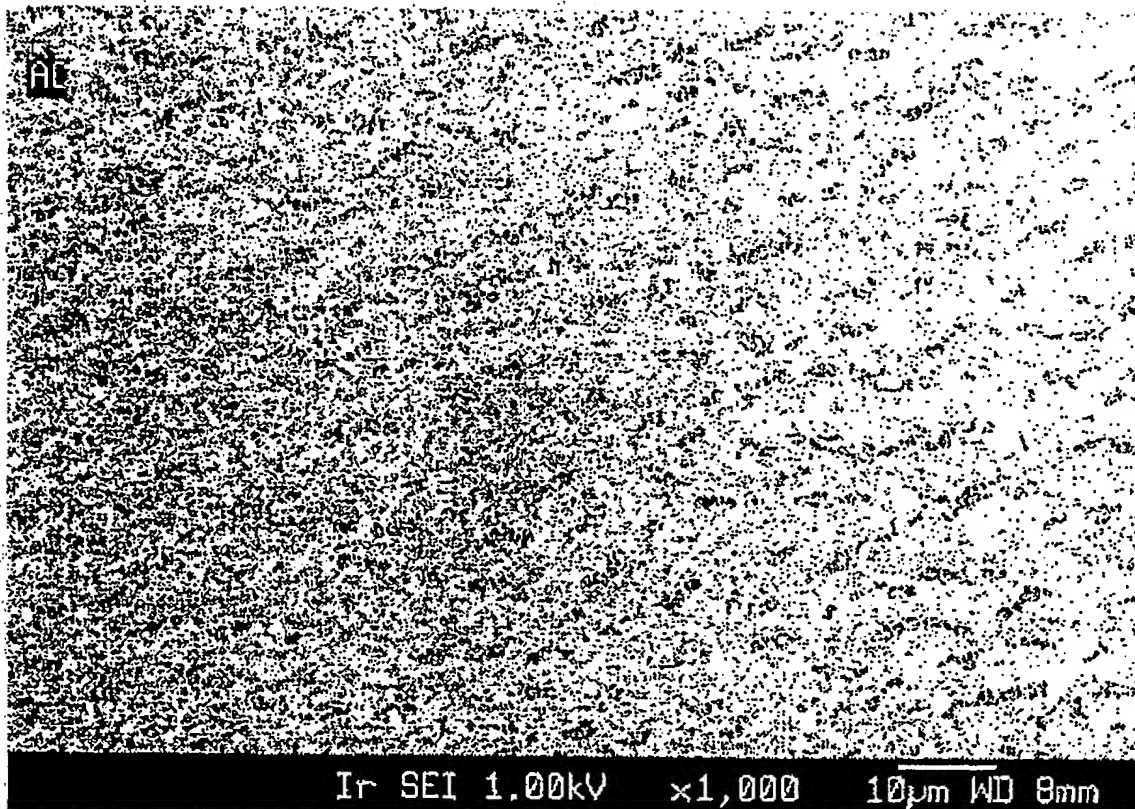
27. The preparation method according to claim 17, wherein an irradiation of step c) is performed under a gas atmosphere selected from the group consisting of helium, hydrogen, nitrogen, ammonia, carbon monoxide,
25 carbon dioxide, chlorofluoro methane, methane, and N_2O atmospheres; and a mixture thereof.

28. The preparation method according to claim 27, wherein the flow rate of the reactive gas ranges from 0.5 to 20 ml/minute.

29. The preparation method according to claim 17, wherein the
30 preparation steps of the multi-component composite membrane can be performed in any combination regardless of the sequence.

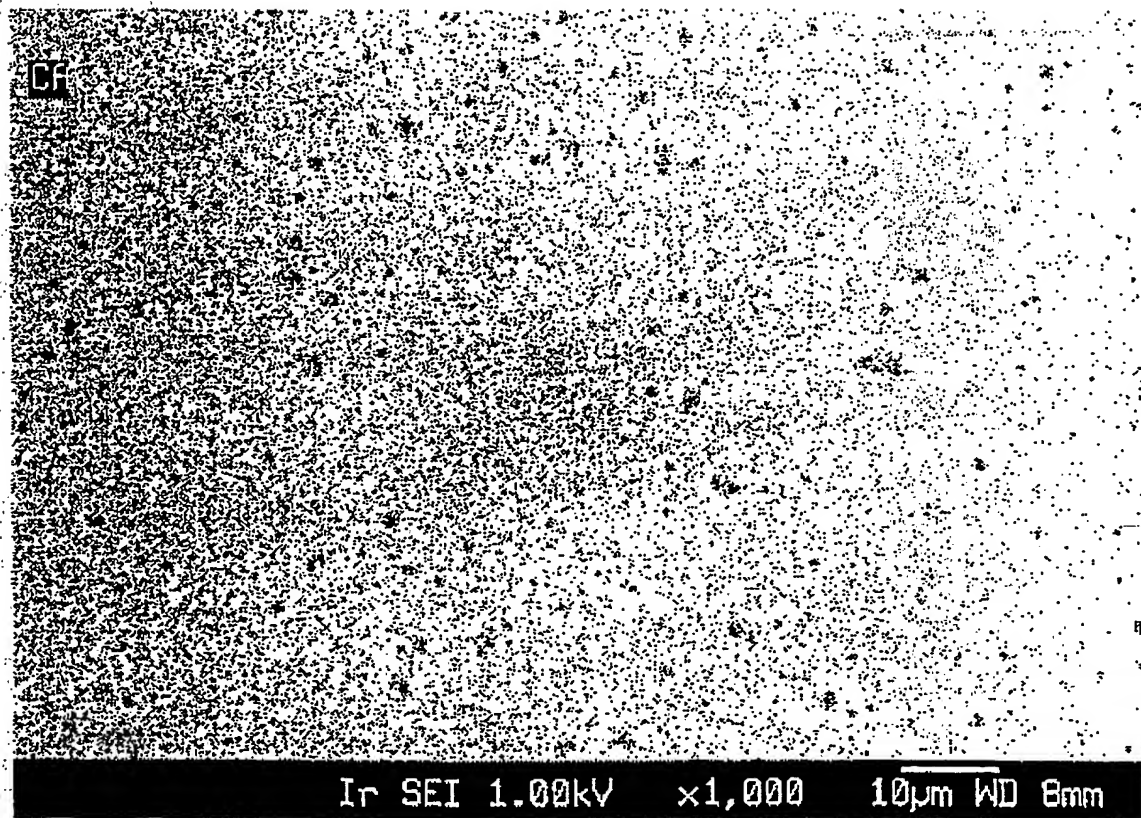
1/2

FIG. 1



2/2

FIG.2





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INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR01/01076

A. CLASSIFICATION OF SUBJECT MATTER**IPC7 B01D 71/06**

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 B01D, B29D, H01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP 10-151330 A (MITSUBISHI PLASTICS IND LTD.) 09. JUNE. 1998. See the whole document	1-29
A	US 5,955,215 A (Kurzweil et al.) 21. SEPTEMBER. 1999. See the whole document	1-29
A	US 4,902,424 A (Wrasidlo) 20. FEBRUARY. 1990. See the whole document	1-29
A	JP 8-281085 A (TOYOCO CO LTD.) 29. OCTOBER. 1996. See the whole document	1, 17-29

☐ Further documents are listed in the continuation of Box C.☒ See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

18 OCTOBER 2001 (18.10.2001)

Date of mailing of the international search report

23 OCTOBER 2001 (23.10.2001)

Name and mailing address of the ISA/KR

Korean Intellectual Property Office
Government Complex-Daejeon, Dunsan-dong, Seo-gu, Daejeon
Metropolitan City 302-701, Republic of Korea

Facsimile No. 82-42-472-7140

Authorized officer

CHAE, Heui Gak

Telephone No. 82-42-481-5565



INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/KR01/01076

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
JP 10-151330 A	09-06-1998	NONE	
US 5,955,215 A	21-09-1999	NONE	
US 4,902,424 A	20-02-1990	NONE	
JP 8-281085 A	29-10-1996	US 5,783,079 A FR 2,723,856 A1	21-07-1998 01-03-1996

1/4

PCT REQUEST

OPP007089KR

Original (for SUBMISSION) - printed on 22.06.2001 05:19:17 PM

0	For receiving Office use only	
0-1	International Application No.	
0-2	International Filing Date	
0-3	Name of receiving Office and "PCT International Application"	
0-4	Form - PCT/RO/101 PCT Request	
0-4-1	Prepared using	PCT-EASY Version 2.91 (updated 01.01.2001)
0-5	Petition The undersigned requests that the present international application be processed according to the Patent Cooperation Treaty	
0-6	Receiving Office (specified by the applicant)	Korean Industrial Property Office (RO/KR)
0-7	Applicant's or agent's file reference	OPP007089KR
I	Title of invention	MULTI-COMPONENT COMPOSITE MEMBRANE AND METHOD FOR PREPARING THE SAME
II	Applicant	
II-1	This person is:	applicant only
II-2	Applicant for	all designated States except US
II-4	Name	LG CHEMICAL CO., LTD.
II-5	Address:	LG Twin Tower, Yoido-dong 20, Youngdungpo-ku, 150-721 Seoul Republic of Korea
II-6	State of nationality	KR
II-7	State of residence	KR
II-8	Telephone No.	+82 42 866 2070
II-9	Facsimile No.	+82 42 863 2053
III-1	Applicant and/or inventor	
III-1-1	This person is:	applicant and inventor
III-1-2	Applicant for	US only
III-1-4	Name (LAST, First)	LEE, Sang-Young
III-1-5	Address:	LG Apt. 5-105, Doryong-dong 381-42, Yousung-ku, 305-340 Taejeon-city Republic of Korea
III-1-6	State of nationality	KR
III-1-7	State of residence	KR

PCT REQUEST

. Original (for SUBMISSION) - printed on 22.06.2001 05:19:17 PM

III-2	Applicant and/or inventor	
III-2-1	This person is:	applicant and inventor
III-2-2	Applicant for	US only
III-2-4	Name (LAST, First)	AHN, Byeong-In
III-2-5	Address:	LG Apt. 9-301 Doryong-dong 381-42, Yousung-ku, 305-340 Taejeon-city Republic of Korea
III-2-6	State of nationality	KR
III-2-7	State of residence	KR
III-3	Applicant and/or inventor	
III-3-1	This person is:	applicant and inventor
III-3-2	Applicant for	US only
III-3-4	Name (LAST, First)	PARK, Soon-Yong
III-3-5	Address:	LG Apt. 3-516 Doryong-dong 386-1, Yousung-ku, 305-340 Taejeon-city Republic of Korea
III-3-6	State of nationality	KR
III-3-7	State of residence	KR
III-4	Applicant and/or inventor	
III-4-1	This person is:	applicant and inventor
III-4-2	Applicant for	US only
III-4-4	Name (LAST, First)	KYUNG, You-Jin
III-4-5	Address:	14/2, 361-5, Gaebong 3-dong, Guro-ku, 152-815 Seoul Republic of Korea
III-4-6	State of nationality	KR
III-4-7	State of residence	KR
III-5	Applicant and/or inventor	
III-5-1	This person is:	applicant and inventor
III-5-2	Applicant for	US only
III-5-4	Name (LAST, First)	SONG, Heon-Sik
III-5-5	Address:	LG Apt. 9-502, Doryong-dong 381-42, Yousung-ku, 305-340 Taejeon-city Republic of Korea
III-5-6	State of nationality	KR
III-5-7	State of residence	KR

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PCT REQUEST

Original (for SUBMISSION) - printed on 22.06.2001 05:19:17 PM


IV-1	Agent r c mmon representative; r address f r correspondence The person identified below is hereby/has been appointed to act on behalf of the applicant(s) before the competent International Authorities as:	agent
IV-1-1	Name (LAST, First)	SONG, Byeong-Ok
IV-1-2	Address:	9th Fl., Teheran Bldg., Yoksam-dong 825-33, Kangnam-ku, 135-080 Seoul Republic of Korea
IV-1-3	Telephone No.	+82 2 3458 0800
IV-1-4	Facsimile No.	+82 2 553 5254
IV-1-5	e-mail	email@mutualip.com
V	Designation of States	
V-1	Regional Patent (other kinds of protection or treatment, if any, are specified between parentheses after the designation(s) concerned)	EP: AT BE CH&LI CY DE DK ES FI FR GB GR IE IT LU MC NL PT SE TR and any other State which is a Contracting State of the European Patent Convention and of the PCT
V-2	National Patent (other kinds of protection or treatment, if any, are specified between parentheses after the designation(s) concerned)	CN JP US
V-5	Precautionary Designation Statement In addition to the designations made under items V-1, V-2 and V-3, the applicant also makes under Rule 4.9(b) all designations which would be permitted under the PCT except any designation(s) of the State(s) indicated under item V-6 below. The applicant declares that those additional designations are subject to confirmation and that any designation which is not confirmed before the expiration of 15 months from the priority date is to be regarded as withdrawn by the applicant at the expiration of that time limit.	
V-6	Exclusion(s) from precautionary designations	NONE
VI-1	Priority claim of earlier national application	
VI-1-1	Filing date	23 June 2000 (23.06.2000)
VI-1-2	Number	2000-34948
VI-1-3	Country	KR
VII-1	International Searching Authority Chosen	Korean Industrial Property Office (KIPO) (ISA/KR)

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PCT REQUEST

OPP007089KR

Original (for **SUBMISSION**) - printed on 22.06.2001 05:19:17 PM

VIII	Check list	number of sheets	electronic file(s) attached
VIII-1	Request	4	-
VIII-2	Description	14	-
VIII-3	Claims	6	-
VIII-4	Abstract	1	EZABST00.TXT
VIII-5	Drawings	2	-
VIII-7	TOTAL	27	
	Accompanying items	paper document(s) attached	electronic file(s) attached
VIII-8	Fee calculation sheet	✓	-
VIII-9	Separate signed power of attorney		-
VIII-16	PCT-EASY diskette	-	diskette
VIII-18	Figure of the drawings which should accompany the abstract	1	
VIII-19	Language of filing of the international application	Korean	
IX-1	Signature of applicant or agent		
IX-1-1	Name (LAST, First)		

FOR RECEIVING OFFICE USE ONLY

10-1	Date of actual receipt of the purported international application	
10-2	Drawings:	
10-2-1	Received	
10-2-2	Not received	
10-3	Corrected date of actual receipt due to later but timely received papers or drawings completing the purported international application	
10-4	Date of timely receipt of the required corrections under PCT Article 11(2)	
10-5	International Searching Authority	ISA/KR
10-6	Transmittal of search copy delayed until search fee is paid	

FOR INTERNATIONAL BUREAU USE ONLY

11-1	Date of receipt of the record copy by the International Bureau	
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PCT (ANNEX - FEE CALCULATION SHEET)

OPP007089KR

Original (for SUBMISSION) - printed on 22.06.2001 05:19:17 PM

(This sheet is not part of and does not count as a sheet of the international application)

0	For receiving Office use only			
0-1	International Application No.			
0-2	Date stamp of the receiving Office			
0-4	Form - PCT/RO/101 (Annex)			
0-4-1	PCT Fee Calculation Sheet Prepared using	PCT-EASY Version 2.91 (updated 01.01.2001)		
0-9	Applicant's or agent's file reference	OPP007089KR		
2	Applicant	LG CHEMICAL CO., LTD., et al.		
12	Calculation of prescribed fees	fee amount/multiplier	total amounts (KRW)	
12-1	Transmittal fee T	⇒	45,000	
12-2	Search fee S	⇒	150,000	
12-3	International fee			
	Basic fee			
	(first 30 sheets) b1	500,000		
12-4	Remaining sheets	0		
12-5	Additional amount (X)	12,000		
12-6	Total additional amount b2	0		
12-7	b1 + b2 = B	500,000		
12-8	Designation fees			
	Number of designations contained in international application	4		
12-9	Number of designation fees payable (maximum 6)	4		
12-10	Amount of designation fee (X)	108,000		
12-11	Total designation fees D	432,000		
12-12	PCT-EASY fee reduction R	-154,000		
12-13	Total International fee (B+D-R) I	⇒	778,000	
12-17	TOTAL FEES PAYABLE (T+S+I+P)	⇒	973,000	
12-19	Mode of payment	cash		

VALIDATION LOG AND REMARKS

13-2-1	Validation messages Request	Green? A translation of the international application into English will have to be prepared under the responsibility of the ISA selected.
		Green? Please note that the ntir request (including the title of invention) must be in English

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PARENT COOPERATION TREATY

PCT

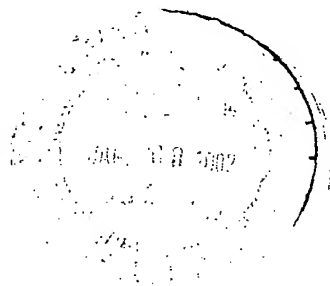
From the INTERNATIONAL BUREAU

NOTICE INFORMING THE APPLICANT OF THE
COMMUNICATION OF THE INTERNATIONAL
APPLICATION TO THE DESIGNATED OFFICES

(PCT Rule 47.1(c), first sentence)

To:

SONG, Byeong-Ok
9th Fl. Teheran Bldg.
Yoksam-dong 825-33
Kangnam-ku
Seoul 135-080
RÉPUBLIQUE DE CORÉE



Date of mailing (day/month/year) 27 December 2001 (27.12.01)		
Applicant's or agent's file reference OPP007089KR		
IMPORTANT NOTICE		
International application No. PCT/KR01/01076	International filing date (day/month/year) 22 June 2001 (22.06.01)	Priority date (day/month/year) 23 June 2000 (23.06.00)
Applicant LG CHEMICAL CO. LTD. et al		

1. Notice is hereby given that the International Bureau has **communicated**, as provided in Article 20, the international application to the following designated Offices on the date indicated above as the date of mailing of this notice:
US

In accordance with Rule 47.1(c), third sentence, those Offices will accept the present notice as conclusive evidence that the communication of the international application has duly taken place on the date of mailing indicated above and no copy of the international application is required to be furnished by the applicant to the designated Office(s).

2. The following designated Offices have waived the requirement for such a communication at this time:
CN, EP, JP

The communication will be made to those Offices only upon their request. Furthermore, those Offices do not require the applicant to furnish a copy of the international application (Rule 49.1(a-bis)).

3. Enclosed with this notice is a copy of the international application as published by the International Bureau on 27 December 2001 (27.12.01) under No. WO 01/97957

REMINDER REGARDING CHAPTER II (Article 31(2)(a) and Rule 54.2)

If the applicant wishes to postpone entry into the national phase until 30 months (or later in some Offices) from the priority date, a **demand for international preliminary examination** must be filed with the competent International Preliminary Examining Authority before the expiration of 19 months from the priority date.

It is the applicant's sole responsibility to monitor the 19-month time limit.

Note that only an applicant who is a national or resident of a PCT Contracting State which is bound by Chapter II has the right to file a demand for international preliminary examination (at present, all PCT Contracting States are bound by Chapter II).

REMINDER REGARDING ENTRY INTO THE NATIONAL PHASE (Article 22 or 39(1))

If the applicant wishes to proceed with the international application in the **national phase**, he must, within 20 months or 30 months, or later in some Offices, perform the acts referred to therein before each designated or elected Office.

For further important information on the time limits and acts to be performed for entering the national phase, see the Annex to Form PCT/IB/301 (Notification of Receipt of Record Copy) and the PCT Applicant's Guide, Volume II.

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland	Authorized officer J. Zahra
Facsimile No. (41-22) 740.14.35	Telephone No. (41-22) 338.91.11

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특 허 협 력 조 약
(PATENT COOPERATION TREATY)

발신:수리관청

수신:

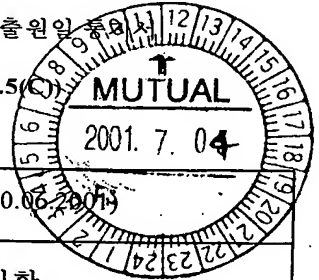
SONG, Byeong Ok

9th Fl., Teheran Bldg., 825-33, Yeoksam-dong,
Kangnam-gu, Seoul 135-080, Republic of KOREA

PCT

국제출원번호 및 국제출원일

(PCT 규칙 20.5(c))

발송일
(일/월/년)

30 JUNE 2001 (30.06.2001)

출원인 또는 대리인의 서류참조기호

OPP007089KR

중요통지사항

국제출원번호

PCT/KR01/01076

국제출원일 (일/월/년)

22 JUNE 2001 (22.06.2001)

우선일 (일/월/년)

23 JUNE 2000 (23.06.2000)

출원인

LG CHEMICAL CO., LTD. et al

발명의 명칭

MULTI-COMPONENT COMPOSITE MEMBRANE AND METHOD FOR PREPARING THE SAME

1. 이 국제출원은 위에 기재된 바와 같이 국제출원번호와 국제출원일이 부여되었습니다.

2. 이 국제출원의 기록원본은:

☐ _____ 자로 국제사무국에 송부되었습니다.

☐ 아래의 사유로 인하여 아직 국제사무국에 송부되지 않았으며, 이 통지서의 사본은 국제사무국에 송부되었습니다*:

☐ 국가안전보장에 필요한 허가를 얻지 못했음

☐ 기타 (이유를 상술):

* 국제사무국은 수리관청에 의한 기록원본의 송달을 감시하고 그 접수사실을 출원인에게 통지합니다.
(서식 PCT/IB/301). 국제사무국은 우선일부터 14월이 경과할 때까지 기록원본을 수령하지 않은 때에는 출원인에게 이를 통지합니다. (규칙 22.1(c)).

수리관청명칭 및 우편주소

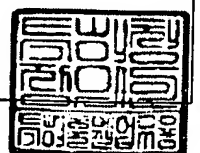
Korean Intellectual Property Office
Government Complex-Daejeon, Dunsan-dong, Seo-gu,
Daejeon Metropolitan City 302-701, Republic of Korea

팩스번호: 82-42-472-7140

특허청장

COMMISSIONER

전화번호: 82-42-481-5206



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PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference OPP007089KR	FOR FURTHER ACTION see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. PCT/KR01/01076	International filing date (<i>day/month/year</i>) 22 JUNE 2001 (22.06.2001)	(Earliest) Priority Date (<i>day/month/year</i>) 23 JUNE 2000 (23.06.2000)
Applicant LG CHEMICAL CO., LTD. et al		

This International search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 3 sheets.

☐ It is also accompanied by a copy of each prior art document cited in this report.

1. Basis of the report

- a. With regard to the language, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

- b. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international search was carried out on the basis of the sequence listing:

☐ contained in the international application in written form.

☐ filed together with the international application in computer readable form.

☐ furnished subsequently to this Authority in written form.

☐ furnished subsequently to this Authority in computer readable form.

☐ the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

☐ the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

2. ☐ Certain claims were found unsearchable (See Box I).

3. ☐ Unity of invention is lacking (See Box II).

4. With regard to the title,

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

5. With regard to the abstract,

☒ the text is approved as submitted by the applicant.

☐ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the drawing to be published with the abstract is Figure No. 1

☒ as suggested by the applicant.

☐ because the applicant failed to suggest a figure.

☐ because this figure better characterizes the invention.

☐ None of the figures.

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A. CLASSIFICATION OF SUBJECT MATTER**IPC7 B01D 71/06**

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 B01D, B29D, H01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP 10-151330 A (MITSUBISHI PLASTICS IND LTD.) 09. JUNE. 1998. See the whole document	1-29
A	US 5,955,215 A (Kurzweil et al.) 21. SEPTEMBER. 1999. See the whole document	1-29
A	US 4,902,424 A (Wrasidlo) 20. FEBRUARY. 1990. See the whole document	1-29
A	JP 8-281085 A (TOYOBO CO LTD.) 29. OCTOBER. 1996. See the whole document	1, 17-29

☐ Further documents are listed in the continuation of Box C.☒ See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

18 OCTOBER 2001 (18.10.2001)

Date of mailing of the international search report

23 OCTOBER 2001 (23.10.2001)

Name and mailing address of the ISA/KR

Korean Intellectual Property Office
Government Complex-Daejeon, Dunsan-dong, Seo-gu, Daejeon
Metropolitan City 302-701, Republic of Korea

Facsimile No. 82-42-472-7140

Authorized officer

CHAE, Heui Gak

Telephone No. 82-42-481-5565



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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/KR01/01076

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
JP 10-151330 A	09-06-1998	NONE	
US 5,955,215 A	21-09-1999	NONE	
US 4,902,424 A	20-02-1990	NONE	
JP 8-281085 A	29-10-1996	US 5,783,079 A FR 2,723,856 A1	21-07-1998 01-03-1996

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